Application No.: 10/574,844

Art Unit: 1795

Amendment under 37 CFR §1.116

Attorney Docket No.: 062327

**REMARKS** 

Rejections under 35 USC §102(b)

Claim 1 was rejected under 35 USC §102(b) as being anticipated by Matsubara et

al. (Matsubara et al., "Fabrication of an all-oxide thermoelectric power generator,"

Applied Physics Letters Vol. 78 (2001): 3627-3629).

Claim 1 has been cancelled. Thus, the rejection has become moot.

Rejections under 35 USC §103(a)

Claims 1-7 were rejected under 35 USC §103(a) as being obvious over Alexander

(U.S. Patent No. 5,422,190) combined with Matsubara et al. (Matsubara et al., "Fabrication

of an All-Oxide Thermoelectric Power Generator," Applied Physics Letters Vol. 78 (2001):

3627-3629).

Claims 1-3 have been cancelled. Thus, the rejection of claims 1-3 has become moot.

As to the rejection of claims 4-7, these claims are directed to an electrically conductive

paste for connecting a p-type thermoelectric material comprising a specific powdery oxide; and at

least one powdery electrically conductive metal selected from the group consisting of gold,

silver, platinum, and alloys containing at least one of these metals. Due to the use of the paste

for connecting thermoelectric materials, a suitable conductivity is given to the connecting portion

of the thermoelectric element, and the separation at the connecting portion can be prevented even

under repeated high-temperature power generation. This makes it possible to maintain good

thermoelectric performance over a long period of time. The Examiner alleged as follows:

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MATSUBARA et al teaches a thermoelectric p-type oxide in the abstract Ca3Co409. This corresponds to the material in formula a in the instant application claim 1. If subscript b and d of the instant application were found to be zero, the material of MATSUBARA et al matches that required in the instant application.

ALEXANDER teaches a via fill paste for use in electronics. The via fill paste is stated, in column 1, lines 27-30, "to provide an electrical bridge or connection between the conductive layers". In the instant application, the conductive layers are the n-type and p-type semiconductors to be connected via the conductive paste, or in ALEXANDER the fill paste. The fill paste is said to be made of "gold, silver and palladium and a refractory oxide", where the oxide simply comprises one or more of a list of metals including the lanthanides. Column 3, lines 25-42, repeatedly prescribe the use of metal powders in the paste.

At the time of the invention, it would have been obvious to one of ordinary skill in the art to combine the oxide thermoelectric material of MATSUBARA et al in the thermoelectric conductive paste of ALEXANDER requiring a conductive oxide because as shown in figure 1 of MATSUBARA et al, the oxide material (comprising the p-leg) conducts electricity from one fin to the other, making it an obvious addition to a conductive paste. Also, it would have been obvious to one of ordinary skill in the art to add a metal, as disclosed in ALEXANDER, to the oxide of MATSUBARA et al to create a conductive paste, as metal is obviously used when electrical conduction is a desirable characteristic.

(Office Action, page 4, last 6 lines to page 5, line 12).

However, the paste of Alexander is a via fill paste for use in the construction of electronic circuit devices, and has nothing to do with a thermoelectric material. Even though the Alexander paste requires conductivity, Alexander discusses the solution of the problem in connecting a layer of conductive silver to a layer of conductive gold. Therefore, nothing in the Alexander indicates that the paste is suitable for a purpose of the present invention, i.e., connecting a thermoelectric material that includes oxides.

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Additionally, Alexander simply discloses "a via fill paste including gold, silver, palladium, and a refractory oxide" as a paste suitable for the aforementioned purpose. Further, although Alexander discloses as a refractory oxide an oxide containing metals such as "zirconium, yttrium . . . and lutetium", it does not disclose complex oxides as recited in claim 4.

In this regard, the Examiner cited Matsubara et al., alleging that the Abstract describes a thermoelectric p-type oxide represented by Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub>, and that such an oxide corresponds to the material defined in claim 1. The Examiner further alleged that Matsubara et al. teaches the addition of a metal to form a paste. Matsubara discloses fabrication of an all-oxide thermoelectric power generator. Matsubara describes in the abstract as follows:

An oxide thermoelectric device was fabricated using Gd-doped Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub> p-type legs and La-doped CaMnO<sub>3</sub> n-type legs on a fin. The power factors of p legs and n legs were  $4.8 \times 10^{-4}$  Wm<sup>-1</sup>K<sup>-2</sup> and  $2.2 \times 10^{-4}$  Wm<sup>-1</sup>K<sup>-2</sup> at 700°C in air, respectively. With eight p-n couples the device generated an output power of 63.5 mW under the thermal condition of hot side temperature Th=773°C and a temperature difference  $\Delta T$ =390°C. This device proved to be operable for more than two weeks in air showing high durability.

(Matsubara, p.3627, abstract). Also, Matsubara describes on FIG.1 as follows:

FIG. 1. Schematic illustration of the fin-type thermoelectric device. Spark plasma sintered  $Ca_{2.75}Ga_{0.25}Co_4O_9$  and conventionally sintered  $Ca_{0.92}La_{0.08}MnO_3$  are used for p and n legs, respectively.

(Matsubara, description of FIG. 1). Thus, in the portions cited by the Examiner, Matsubara discusses Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub> as **p-type legs** of the thermoelectric device, but not as material for a connecting oxide.

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As mentioned above, Alexander discloses a via fill paste for use in the manufacture of

electronic circuit devices, which has nothing to do with the thermoelectric material. Even though

the paste requires conductivity, the Alexander's paste is used to solve the problem of connecting

a layer of conductive silver to a layer of conductive gold, which is completely different from that

of the present invention, i.e., connecting a thermoelectric material.

Thus, there is no reason to combine Alexander and Matsubara et al. to produce a paste for

the specific purpose, i.e., for connecting thermoelectric materials. In particular, the following

effect of the invention would not have been expected from the combined teachings of the

References, i.e., the effect that, by using the specific oxide-containing paste not only conductivity

is imparted to the connecting portion, but favourable performance is also maintained at the

connecting portion without causing separation, even when the thermoelectric material is

repeatedly used at high temperatures.

Moreover, Matsubara et al. does not disclose the complex oxide, which is contained in

the paste recited in claim 4. Although the Examiner alleged that Matsubara et al. teaches

Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub> Matsubara et al. does not disclose Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub> but it actually discloses Gd-doped

Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub> in the Abstract, and Ca<sub>3</sub>Gd<sub>0.25</sub>Co<sub>4</sub>O<sub>9</sub> as the specific composition in Table 1. Claim 4

recites that, in Ca<sub>2</sub>A<sup>1</sup><sub>b</sub>Co<sub>c</sub>A<sup>2</sup><sub>d</sub>O<sub>e</sub>, A<sup>1</sup> is "one or more elements selected from the group consisting

of Na, K, Li, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Pb, Sr, Ba, Al, and Bi". Thus, the complex oxide

contained in the paste of Claim 4 does not include Gd, which is contained in the oxide of

Matsubara et al.

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The Examiner is required to give a reason why a person of ordinary skill in the art would

combine or modify the prior art references to result in the claimed invention. It is not logical that

it would have been obvious to combine the oxide thermoelectric material of Matsubara et al. with

the thermoelectric conductive paste (actually "via fill paste") of Alexander requiring a conductive

oxide because the oxide material (comprising the p-leg) conducts electricity from one fin to the

other, making it an obvious addition to a conductive paste because the via fill paste has nothing

to do with the thermoelectric material. There is no reason to combine the via fill paste disclosed

in Alexander and the Ca<sub>3</sub>Co<sub>4</sub>O<sub>9</sub> used as p-type legs of the thermoelectric device, which is

disclosed in Matsubara.

Thus, because the rejection has not given any valid rationale to support a conclusion of

obviousness, a prima facie case of obviousness has not been established.

The present invention was made under the following background:

The realization of efficient thermoelectric generation requires a thermoelectric element

comprising a pair of connected p-type and n-type thermoelectric materials, and a thermoelectric

module obtained by integrating thermoelectric elements, i.e., an electric power generator.

However, the development of thermoelectric elements and thermoelectric modules has been

delayed when compared with the development of thermoelectric materials.

In particular, the development of a method for connecting thermoelectric materials with a

low electrical resistance is important for putting thermoelectric modules into practical use. In the

case of thermoelectric generation using high-temperature waste heat of 673 K (400°C) or higher,

thermoelectric materials are connected using, as a binder, a paste containing a noble metal such

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as silver, gold, or platinum because a connecting portion formed by soldering is likely to oxidize

or melt under such conditions.

However, such noble metal pastes are not suitable when oxides are used as substrate

materials, thermoelectric material, etc. because there is a large difference in the thermal

expansion coefficient between the oxide and the noble metal contained in the paste. Thus,

repeated high-temperature power generations cause separation at the connecting portion,

resulting in increased internal resistance and lowered mechanical strength. The connecting

portion therebetween also has a problem of a large interface resistance due to contact between the

metal and oxide.

The present invention solves the above problems. The present invention provides a

material for connecting thermoelectric materials which can achieve the connection of the

thermoelectric material made of oxide with a low electrical resistance and which hardly arises a

performance deterioration of a thermoelectric module even when repeating high-temperature

power generation and further to provide a thermoelectric element produced using such a material

for connecting thermoelectric materials.

Nothing in Alexander and Matsubara indicates that the claimed electrically conductive

paste can give excellent performance in a thermoelectric module.

For at least these reasons, claim 4 patentably distinguishes over Alexander and

Matsubara. Claims 5-7, depending from claim 4, also patentably distinguish over Alexander and

Matsubara for at least the same reasons.

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Claims 8-11 were rejected under 35 USC §103(a) as being obvious over Alexander

(U.S. Patent No. 5,422,190) combined with Funahashi et al. (JP Abstract Publication

number 2003-282964).

Claims 8-11 have been cancelled. Thus, this rejection has become moot.

Claims 12-14 were rejected under 35 USC §103(a) as being obvious over Alexander

(U.S. Patent No. 5,422,190) combined with Matsubara et al. (Matsubara et al., "Fabrication

of an all-oxide thermoelectric power generator," Applied Physics Letters Vol. 78 (2001):

3627-3629) and Funahashi et al. (JP Abstract Publication number 2003-282964).

Claim 12 has been cancelled. Thus, the rejection of claim 12 has become moot.

Funahashi discloses a complex oxide having high Seebeck coefficient and high electric

conductivity. Funahashi discusses nothing about connecting material. Thus, Funahashi does not

remedy the deficiencies of Alexander and Matsubara et al.

Claims 13 and 14 relate to a thermoelectric element wherein one end of a p-type

thermoelectric material and one end of an n-type thermoelectric material are each connected to an

electrically conductive substrate with an electrically conductive paste.

Claim 13 specifically defines the compositions of the p-type and n-type thermoelectric

materials, and the paste having the same composition as recited in claim 4 is used for

connecting the p-type thermoelectric material. Claim 14 recites that a paste having the same

composition as recited in claim 5 is used for connecting the p-type thermoelectric material.

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Therefore, for at least the reasons as discussed regarding claim 4, claims 13 and 14 also

patentably distinguish over Alexander, Matsubara and Funahashi.

Claims 15 and 16 were rejected under 35 USC §103(a) as being obvious over

Alexander (U.S. Patent No. 5,422,190) combined with Matsubara et al. (Matsubara et al.,

"Fabrication of an all-oxide thermoelectric power generator," Applied Physics Letters Vol.

78 (2001): 3627-3629) and Funahashi et al. (JP Abstract Publication number 2003-282964)

as applied to claim 12 above, and further in view of Buist (U.S. Patent No. 4,859,250).

Claims 15 and 16 have been cancelled. Thus, this rejection has become moot.

Claims 17 and 18 are rejected under 35 USC §103(a) as being obvious over

Alexander (U.S. Patent No. 5,422,190) combined with Matsubara et al. (Matsubara et al.,

"Fabrication of an all-oxide thermoelectric power generator." Applied Physics Letters Vol.

78 (2001): 3627-3629) and Funahashi et al. (JP Abstract Publication number 2003-282964)

as applied to claim 13 above, and further in view of Buist (U.S. Patent No. 4,859,250).

Claim 17 is directed to a thermoelectric module comprising a plurality of the

thermoelectric elements of claim 13, and claim 18 is directed to a thermoelectric conversion

method using the module.

With regard to these claims, the Examiner cites Buist (U.S.P. No. 4,859,250), in addition

to the aforementioned references. Buist appears to disclose a device in which the elements are

connected in a similar manner to that defined in claim 17 (see Fig. 3a), and a method for power

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generation using a positioning process similar to that defined in claim 18 (see Fig. 4). Claim 17,

however, further recites a plurality of the thermoelectric elements of claim 13. As discussed

above, these thermoelectric elements are unobvious over any combination of the aforementioned

references.

For at least these reasons, claims 17 and 18 patentably distinguish over Alexander,

Matsubara, Funahashi and Buist.

In view of the aforementioned amendments and accompanying remarks, Applicants

submit that the claims, as herein amended, are in condition for allowance. Applicants request

such action at an early date.

If the Examiner believes that this application is not now in condition for allowance, the

Examiner is requested to contact Applicants' undersigned attorney to arrange for an interview to

expedite the disposition of this case.

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If this paper is not timely filed, Applicants respectfully petition for an appropriate extension of time. The fees for such an extension or any other fees that may be due with respect to this paper may be charged to Deposit Account No. 50-2866.

Respectfully submitted,

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